

# Internal laser cooling of $\text{Yb}^{3+}$ -doped glass measured between 100 and 300 K

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Laser cooling of a solid can occur when it emits photons of higher mean energy than those it absorbs. Photothermal deflection spectra of a fluorozirconate glass (ZBLANP) doped with 1 wt%  $\text{Yb}^{3+}$  show cooling in an internal volume of the sample at temperatures between 100 and 300 K. A cooling efficiency of  $\sim 1\%$  relative to the absorbed laser power at 1015 nm is maintained at all temperatures. The results show promise for solid-state cooling of bulk materials at temperatures below 150 K. © 1997 American Institute of Physics. [S0003-6951(97)04937-1]

In 1950, the French spectroscopist A. Kastler proposed that an *effet lumino-frigorifique*—optical cooling—of rare-earth-doped crystals might result from anti-Stokes emission.<sup>1</sup> In 1968, the first reported attempt to experimentally cool a solid with light was performed—the  $1.064\text{ }\mu\text{m}$  transition of  $\text{Nd}^{3+}$ :YAG was pumped using a laser of the same composition.<sup>2</sup> In this experiment, only reduced heating of the sample was observed; contamination by  $\sim 50$  ppb of  $\text{Dy}^{3+}$  was thought to produce enough residual heating to overwhelm the cooling. Reduced heating has also been observed<sup>3</sup> for laser-cooled GaAs and net cooling for solvated laser dyes.<sup>4–6</sup> It is only recently that net optical cooling of a solid was observed,<sup>7</sup> namely for a rectangular piece of  $\text{Yb}^{3+}$ -doped ZBLANP ( $\text{ZrF}_4\text{-BaF}_2\text{-LaF}_3\text{-AlF}_3\text{-NaF-PbF}_2$ ) glass. This host material can be carefully purified using techniques developed for telecommunications applications.<sup>8</sup> External cooling efficiencies of up to 2% were measured at room temperature,<sup>7</sup> large enough to merit consideration for practical solid-state refrigeration of bulk materials. To determine the feasibility of this proposal, measurements and modeling of the cooling of a localized volume of the interior of a sample (avoiding possible heating from surface impurities or from reabsorption of the emitted light) as a function of temperature are a necessary first step.

In this Letter, the laser-cooling efficiencies of a bulk sample of ZBLANP + 1 wt%  $\text{Yb}^{3+}$  are characterized over a range of pump wavelengths and temperatures. The term “efficiency” here refers to the ratio of the cooling power to the absorbed laser power (to be distinguished from the incident laser power). Photothermal deflection spectroscopy is employed, which is a technique originally developed<sup>9</sup> in 1980 as a means of measuring optical absorbances down to  $10^{-8}$  and which has since become a well-established spectroscopic tool.<sup>10</sup> An optical probe beam is deflected from a refractive-index gradient set up in a locally heated (or cooled) volume of the sample. The temperature change is produced by focusing a pump beam on it which is partially absorbed by the sample. We find that the ratio of the cooling power to the absorbed laser power is  $\sim 1\%$  even for temperatures as low as 100 K.

The experimental setup is sketched in Fig. 1. An argon-ion-pumped titanium-sapphire laser is tunable over the range 900–1050 nm, spanning the absorption spectrum<sup>7</sup> of ZBLANP: $\text{Yb}^{3+}$ . A counter-propagating helium-neon probe laser beam, at a wavelength of 632.8 nm where the sample is transparent, is co-aligned with the pump beam. This HeNe laser is temperature controlled to reduce pointing fluctuations and optically isolated to prevent feedback. Dichroic beam splitters, which are highly reflecting at  $1\text{ }\mu\text{m}$  and transmitting at 632.8 nm, are used to combine the pump and probe beams, each of which is focused into the same volume in the sample. The pump is chopped at 0.6 Hz and the probe beam is synchronously measured with a position-sensitive detector. The HeNe beam is displaced laterally by  $\sim 50\text{ }\mu\text{m}$  from the pump beam in the sample, so as to most effectively probe the thermal gradient. The resulting signal is filtered to reduce high-frequency environmental noise. When the chopper shutter either opens or closes, an approximately exponential deflection signal is observed, with a  $\sim 50$  ms time constant, consistent with the thermal diffusivity of ZBLANP. (The thermal time constant of the entire sample is much longer than the rapid temperature changes induced in the small volume of the sample illuminated by the pump laser.) Care must be taken to eliminate, by judicious choice of the lateral dis-

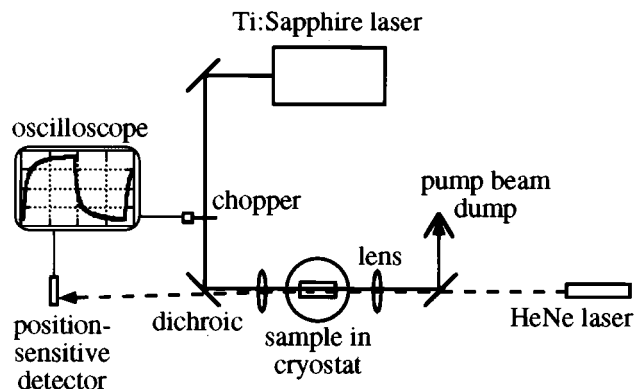


FIG. 1. Experimental setup for photothermal deflection spectroscopy. A tunable cw Ti:Sapphire laser is chopped and focused into the sample to produce either heating or cooling in the approximately cylindrical volume of the beam waist. A counter-propagating HeNe laser beam is deflected by the thermally induced gradient in the refractive index and is synchronously detected using a position-sensitive detector.

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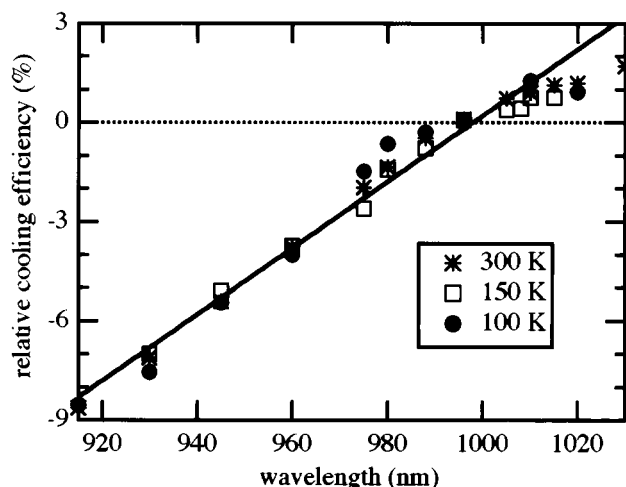


FIG. 2. Relative cooling efficiencies as a function of the laser pump wavelength. The solid line is the theoretical prediction for a mean fluorescence wavelength of 998 nm, and the symbols give the experimental data at the indicated temperatures. The data have been scaled to match the theoretical curve at around 960 nm where the heating is large and is dominated by the well-characterized  $\text{Yb}^{3+}$  absorption and fluorescence.

placement, rapid transients in the signal which can arise from the finite time required for the chopper blades to cross the  $\sim 2$ -mm pump beam diameter. The sample is suspended in an optical-access helium-gas-immersion cryostat.

The peak-to-peak amplitude and the phase in the deflection signal are measured as a function of the pump wavelength for each bath temperature. The magnitude of the deflection angle is proportional to the radial gradient in the refractive index and hence in the local temperature. The phase of the deflection, relative to that of the pump beam, changes by  $180^\circ$  during the transition from laser-induced heating to cooling because the temperature change reverses sign.<sup>11</sup> The measured amplitude is proportional to the laser-induced temperature change,  $\Delta T$ , at the center of the pumped volume. Since this temperature change is small relative to the cryostat bath temperature,  $\Delta T$  is approximately proportional in turn to the optical cooling power,  $P_{cool}$ . Normalizing this cooling power by the absorbed laser power,  $P_{abs} = P_{inc}[1 - \exp(-\alpha L)]$ , defines the relative cooling efficiency,  $\eta_R = P_{cool}/P_{abs}$ . Here  $P_{inc}$  is the incident laser power, while  $\alpha$  and  $L$  are the absorption coefficient and length of the sample, respectively. (The absorption spectra at the temperatures of interest are graphed and discussed in Ref. 12.) Photothermal deflection measurements thus determine the relative cooling efficiency to within a proportionality factor. The results are plotted in Fig. 2 using the normalization discussed in the following paragraph.

The proportionality factor depends on various material parameters, the sample temperature, and the experimental setup. At each temperature, the overall factor can be determined by comparison with theory. Energy conservation requires that the cooling rate is equal to the difference between the radiant power that enters and leaves the sample. This implies that the relative cooling efficiency is  $\eta_R = (\lambda - \lambda_F^*)/\lambda_F^*$ , where  $\lambda$  is the laser pump wavelength and  $\lambda_F^* \equiv \lambda_F/\eta$ . Here  $\lambda_F$  is the wavelength corresponding to the mean energy of the emitted photons that escape and  $\eta$  is the

quantum efficiency for photon emission and escape. At each temperature, the data show that  $\eta_R$  varies approximately linearly with  $\lambda$ , as predicted. The transition wavelength  $\lambda_F^*$  between heating and cooling is experimentally determined by the  $180^\circ$  phase shift in the photothermal deflection signal relative to the modulated pump beam. A comparison of  $\lambda_F^*$  with  $\lambda_F$  from the measured fluorescence spectra<sup>12</sup> shows that the quantum efficiency  $\eta$  is not measurably different from unity at any temperature. Specifically, as the sample temperature is decreased from 300 to 100 K, the fluorescent spectrum changes shape and the value of  $\lambda_F$  increases from 995 to 1001 nm.<sup>12</sup> The theoretical curve in Fig. 2 uses the average value of 998 nm for simplicity.

If the quantum efficiencies were less than unity, the experimental data would cross zero at longer wavelengths than  $\lambda_F$  due to heating effects which scale with the ytterbium absorption.<sup>13</sup> The intrinsic nonradiative decay of the excited ytterbium ions is negligible, owing to the large energy gap between the  $^2F_{5/2}$  and  $^2F_{7/2}$  multiplets. Heating, by energy transfer from excited  $\text{Yb}^{3+}$  ions to impurities having nearly resonant energy levels (such as transition-metal or other rare-earth ions, which can decay nonradiatively), is also minimal due to the high purity of the ZBLANP host. The experimental cooling efficiencies in Fig. 2 fall below the theoretical expectations at  $\lambda > 1015$  nm, however. This roll-off may indicate direct, broadband absorption of the pump laser light by bulk or surface impurities. Such heating would be most evident at long wavelengths, where the  $\text{Yb}^{3+}$  absorption is weakest.

Figure 2 shows that the cooling efficiency of the glass remains in good agreement with theory down to temperatures of at least 100 K. This supports the prospect of practical solid-state refrigeration. In particular, the absolute cooling powers and expected temperature drops of such a process show promise based on the measured fluorescence and absorption spectra.<sup>12</sup>

In conclusion, photothermal deflection spectroscopy demonstrates that an internal volume of a bulk sample of ZBLANP: $\text{Yb}^{3+}$  cools at temperatures between 100 and 300 K. To extend these results to net cooling of the entire sample, it will be necessary to pass the laser pump beam through the solid many times, in order to compensate for the reduction in the absorption coefficient of  $\text{Yb}^{3+}$  at long wavelengths, particularly as the temperature falls.<sup>12</sup> Furthermore, other experiments have indicated that the fluorescence quantum efficiency may be degraded by surface impurities<sup>13</sup> and care must be taken to minimize this effect.

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